

## REPORT DOCUMENTATION PAGE

Public reporting burden for this collection of information is estimated to average 1 hour per response, including maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

Form 298  
and  
Instructions  
April 1989

1. AGENCY USE ONLY (Leave Blank)	2. REPORT DATE August 1995	3. REPORT TYPE AND DATES COVERED Final Technical Report, 7/1/92-6/30/95	
4. TITLE AND SUBTITLE Tunneling Spectroscopy of Ultrasmall Clusters and Grains		5. FUNDING NUMBERS 61103D 3484/S3	
6. AUTHOR(S) K. Likharev		8. PERFORMING ORGANIZATION REPORT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) State University of New York at Stony Brook Stony Brook, NY 11794-3800		10. SPONSORING/MONITORING AGENCY REPORT NUMBER F49620-92-J-0358	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR-NE Dr. Harold Weinstock 110 Duncan Ave. Bolling AFB, DC 20332-0001		11. SUPPLEMENTARY NOTES	
12a. DISTRIBUTION/AVAILABILITY STATEMENT <b>Approved for public release; distribution unlimited.</b>		12b. DISTRIBUTION CODE <b>19951002 018</b>	
13. ABSTRACT (Maximum 200 words) The Final Technical Report describes results obtained during the research project supported by an AFOSR AASERT Grant #F49620-92-J-0358 (parent grant #AFOSR-91-0445). The report consists of 3 sections: 1. Summary 2. Main Results 3. Conclusion and Prospects. The main research achievement during this work has been the development of an original instrument for scanning tunneling microscopy and spectroscopy, with electronic remote control of all its functions (including the coarse approach of the scanning tip), for a broad range of research applications.  DTIC QUALITY INSPECTED 5			
14. SUBJECT TERMS Scanning tunneling microscopy, tunneling spectroscopy, single-electronics, nano-objects, clusters, thin films		15. NUMBER OF PAGES	
17. SECURITY CLASSIFICATION OF REPORT		18. PRICE CODE	
18. SECURITY CLASSIFICATION OF THIS PAGE		20. LIMITATION OF ABSTRACT	
19. SECURITY CLASSIFICATION OF ABSTRACT			

# **TUNNELING SPECTROSCOPY OF ULTRASMALL CLUSTERS AND GRAINS**

**AFOSR Grant # F49620-92-J-0358**  
(FY91 AASERT)  
Parent Grant # AFOSR-91-0445

## **Final Technical Report**

**Principal Investigator:** Prof. Konstantin K. Likharev

**Address:** Department of Physics  
State University of New York  
Stony Brook, NY 11794-3800

Phone: 516-632-8159  
Fax: 516-632-8774  
e-mail: KLIKHAREV@CCMAIL.SUNYSB.EDU

**Project Period:** July 1, 1992 - June 30, 1995

August 1995

## 1. Summary

Single-electronics is a new and exciting field of physical and applied electronics, which has emerged during the last decade. The physics of this field is based on the effects of correlated single-electron tunneling. Their essence is that the transfer of single electrons in systems of conducting (metallic, semiconductor, or molecular) "electrodes", connected by tunnel barriers of very small area, may be strongly correlated either in time, or in space, or both.

Since 1987, reliable evidence of correlated tunneling has been obtained in numerous experiments with normal-metal, superconductor, and semiconductor junctions and systems. Preliminary studies of possible applications have shown that single-electronics may yield a completely new generation of both digital and analog devices with unparalleled performance, most notably extremely dense digital circuits with up to  $10^{11}$  active devices (logic gates and/or memory cells) per square centimeter. However, in order to bring single-electronics to practice, numerous problems must be solved. In 1991, three research groups at SUNY - Stony Brook, headed by Professors Dmitri Averin, Konstantin Likharev and James Lukens, working in collaboration, began an AFOSR-supported project in the field of single-electronics. As a result of this effort, a solid technological, experimental and theoretical base for single-electronics was established at Stony Brook, and several important results have been obtained.

The main objectives of the present supplementary project have been:

- to extend the work to scanning tunneling microscopy/spectroscopy (STM/S) of various nanostructures (ultimately at low temperatures), and
- to increase the number of graduate students involved in research in the field of single-electronics.

During the first two years of the project, the research was carried out in collaboration with the group of Dr. Myron Strongin at the Brookhaven National Laboratory. The collaboration was very pleasant and fruitful, and provided a substantial leverage for our AASERT award.

Our main research achievement has been *the development of an original STM/S instrument with electronic remote control of all its functions (including the coarse approach), with a broad range of possible applications*. This instrument has been used to carry out STM studies of a broad variety of nanostructures, including Moire patterns on graphite surfaces, monolayers of stearic acid on graphite, atomically-smooth gold thin films on mica, monolayers of C<sub>60</sub> buckyballs on atomically-smooth gold surfaces (in particular, modified by extremely thin gold overlayers), and metal-doped diamond-like carbon.

A more detailed description of the main results of this work is given below.

Availability Codes	
Dist	Avail and/or Special
A-1	

## **2. Main Results**

### **A. Simulation of Interaction of Single-Electron Solitons**

For adequate interpretation of anticipated experimental results on single-electron charging of 1D arrays of nano-objects, we have carried out analytical and numerical calculations of interaction of single-electron solitons in such arrays, using simple geometrical models. In contrast to previous calculations which had used oversimplified assumptions, we have found a natural crossover between the exponential decrease of the electrostatic field near the center of the soliton and the virtually unshielded ( $1/r$ ) decrease of the scalar potential at large distances. A simple expression for the stray capacitance per period has been derived from these numerical results [1].

### **B. STM/S Instrument with New Piezoelectric Engine**

For our single-electron nanospectroscopy work we would like to have an STM/S instrument which could operate at helium and millikelvin temperatures and high vacuum without any mechanical links to the ambient environment. (Commercially available low-temperature STMs are not applicable in our experimental conditions). This is why we have designed and implemented a preliminary version of such an STM/S using a new type of piezoelectric engine for the tip position adjustment [2].

The engine consists of a  $60^\circ$  quartz prism (carrying the piezo-tube STM scanner) which can move along a  $60^\circ$  cut in a Macor body of the sample holder. A spring presses the prism against 4 shear-motion piezo plates ("legs") which are glued to the body. During the first stage of each crude approach step, the voltage on all the legs is changed simultaneously. The upper surfaces of the prisms shift along the cut, and carry the prism with them. Then the voltage is removed from the legs one-by-one; during this stage the prism stays in place, because at each instant its friction against 3 static legs outweighs the friction against 1 moving leg.

Our engine, and the STM/S as a whole, operates reliably at room temperatures (both in air and high vacuum) and has been applied to studies of several nanosystems which seemed promising for the observation of correlated single-electron tunneling effects.

### **C. STM/S Studies of Nanostructures**

#### **C.1. Moiré patterns on HOPG**

Highly oriented pyrolytic graphite (HOPG) is a commonly used substrate for STM/S applications. Using our STM/S we could observe atomic patterns on the HOPG surfaces, including the Moiré patterns associated with variations of local stacking [2], without any vibration isolation. We have discovered an extended electronic effect from a single atomic vacancy, and measured the decay factor along the c-axis and various bias voltage dependent topographies for the Moiré patterns [3]. These results may be very useful for further studies of ultrasmall particles and clusters on HOPG substrates.

### **C.2. Macromolecules on HOPG**

To fix ultrasmall conducting particles on a graphite surface, Langmuir-Blodgett films of stearic acid mixed with 1,7-(CH<sub>3</sub>)<sub>2</sub>-1,2-C<sub>2</sub>B<sub>10</sub>HgTl(OCOCF<sub>3</sub>)<sub>2</sub> molecules were prepared using distilled water, and transferred onto freshly cleaved HOPG substrate using the Langmuir-Schäfer method. Our STM study has shown that the surfaces of the graphite substrates had been modified. We have determined that the modification was caused by the solvent intercalation. While being detrimental for our particular attempt to fix organic clusters on the surface, this effect may be useful in general to characterize surface damage caused by different sample preparation techniques.

### **C.3. Au (111) and C<sub>60</sub>**

Gold films were deposited at 440°C on pre-baked mica substrates. Using our STM we could get atomic resolution on these films in air. The STM studies have shown that atomically flat regions on the films were ~0.1 μm wide. By evaporation of buckyballs (C<sub>60</sub>) on the surface we could obtain and image well-ordered single-molecular layers with hexagonal structure [4]. To our surprise, the morphology of these structures changes dramatically after adding one more Au overlayer. While the first C<sub>60</sub> monolayer remained flat, the extra buckyballs formed clusters with an average size of 8 nm.

### **C. 4. Some other applications**

Our STM/S instrument has been used for some purposes beyond the scope of this project (the corresponding research costs were covered from funds different from the AASERT award). For example, the BNL group have used this instrument to study metal-containing diamond-like nanocomposites (a-C:H)/(a-Si:O)/Me films [4]. The electrical and structural properties of these films could be controlled by several means, e.g., metal doping, thermal annealing and probably high field (or heating) in the vicinity of an STM tip. Using STM, the annealing- induced sp<sup>2</sup>-to-sp<sup>3</sup> transformation (which may provide an interesting way to engineer nanostructured samples) has been observed [4].

## **3. Conclusion and Prospects**

Unfortunately, we have not been able to find sufficiently stable objects which could exhibit correlated single-electron tunneling effects at room temperature. On the other hand, low-temperature (~5 K) operation of our instrument was not stable enough for its systematic use. Nevertheless, the instrument has turned out to be extremely useful for several room-temperature studies of issues closely related to the future development of nanoelectronics. Our current plans are to use it mostly for in-situ studies of the thermal growth of very thin (~2-nm) aluminum oxide layers used as tunnel barriers in niobium-trilayer tunnel junctions for our single-electronics research within our parent AFOSR project, as well as for superconductor electronics work funded by DoD's URI through another AFOSR grant.

#### 4. References

1. K.K. Likharev and K. Matsuoka, "Electron-Electron Interaction in Linear Arrays of Small Tunnel Junctions", submitted to Appl. Phys. Lett. (July 1995).
2. Z.Y. Rong and P. Kuiper, "Electronic Effects in Scanning Tunneling Microscopy: Moiré Pattern on a Graphite Surface", Phys. Rev. B **48**, 17427 (1993).
3. Z.Y. Rong, "Extended Modifications of Electronic Structures Caused by Defects: Scanning Tunneling Microscopy of Graphite", Phys. Rev. B **50**, 1839 (1994).
4. Z.Y. Rong and L. Rokhinson, "STM Study of Gold-overlayer Formation on C<sub>60</sub> Monolayers", Phys. Rev. B **49**, 7749 (1994).
5. Z.Y. Rong, M. Abraizov, B. Dorfman, M. Strongin, X.-Q. Yang, D. Yan, and F. H. Pollak, "Scanning Tunneling Microscopy of Diamond-like Nanocomposite Films", Appl. Phys. Lett. **65**, 1379 (1994).